Evaluation of 3M Empore™ Rad Disks for Radium in Water

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Abstract

Empore™ Radium Disks, manufactured by the 3M Corporation, have been evaluated for use in EML's Quality Assurance Program (QAP) for the rapid determination of Radium in Water. These 47 mm diameter by 500 μm thick membrane disks are impregnated with a crown ether that selectively extracts β-emitting ^{225}Ra ($t_{1/2}$ = 14.8 dy) or ^{228}Ra ($t_{1/2}$ = 6.7 yr) and α-emitting ^{223}Ra ($t_{1/2}$ = 11.2 dy), ^{224}Ra ($t_{1/2}$ = 3.6 dy) or ^{226}Ra ($t_{1/2}$ = 1622 yr) from water. Typically, the Ra bearing solution is loaded onto the disk from 2N HNO_3 and eluted with 0.25M (basic) ethylenediamine tetraacetic acid (EDTA). Using 10-20 mL amounts of non-acidified matrix free samples, we pre-conditioned each disk with 2N HNO_3 and individually measured the retention and elution characteristics of 20 isotopes that are constituents of EML's QAP samples. We utilized either (1) Liquid Scintillation Analysis (LSA), (2) NaI(Tl) gamma counting or (3) Solid State Alpha Spectrometry to make our measurements, rather than ^{222}Rn emanation, which is specific only for ^{226}Ra. We found that the disks could be counted directly (for ^{226}Ra) using a low background gas-proportional α/β counter, with 10% α-counting efficiency, or a LSA, with 99% α-counting efficiency. The 6 α-emitting actinides tested were ^{230}Th, nat U, ^{242}Pu, ^{243}Am, ^{237}Np and ^{244}Cm. We selected several β/γ-emitting Group I (^{3}H, ^{40}K, ^{137}Cs) and Group II (^{45}Ca, ^{90}Sr/Y, ^{133}Ba) elements as potential interferents when measuring Radium by LSA. Three transition elements (^{55}Fe, ^{54}Mn, ^{106}Ru/Rh) as well as ^{226}Ra and 2 of its β-emitting progeny, ^{210}Pb and ^{210}Bi were individually tested. We found that ^{210}Po and ^{210}Bi, were not retained by the disk while > 95% of added ^{226}Ra was recovered from samples that we tested. The only other divalent elements that eluted with EDTA were Pb (90% eluted), Sr (85%) and Ba (20%). For three QAP water samples spiked with 4 dpm (0.6 Bq) of ^{226}Ra, the found to added ratio was 0.90 $\pm$ 0.14. The 20 mL QAP samples (acidified with 1N HCl) contained ^{3}H (300 dpm), ^{55}Fe (99 dpm), ^{90}Sr/Y (1.8 dpm) ^{238}Pu (2.2 dpm), ^{241}Am (1 dpm), ^{54}Mn (51 dpm), ^{60}Co (42 dpm) and ^{137}Cs (54 dpm). For rapid screening , good results were obtained following a 15 min LS count of either the unstripped 3M disk or EDTA fraction. Samples containing < 2 dpm (0.03 Bq) were below the LLD of our LS counter and required a direct α-count of ^{226}Ra. This was accomplished by microprecipitating Radium (as $Ra/BaSO_4$) at pH 4-5 from the EDTA fraction, following Sill's method. A first attempt to measure Radium in New York City tap water was unsuccessful because of the presence of gram amounts of soluble NO_3 salts that presumably interfered with ^{226}Ra extraction. Since the Radium concentration in NYC water is extremely low (0.4 mBq L^{-1}), we evaporated 50 L to 100 mL. Prior to pre-filtering with glass-fiber pads, we wet ashed with concentrated HF, HCl and finally HNO_3. Following an 18 h count, we were unable to detect a ^{226}Ra α-peak in the EDTA strip fractions of 4 pre-filtered and concentrated NYC water samples, one of which was internally spiked with 3 dpm of ^{226}Ra. In each case, we accounted for 86-95% of γ-emitting ^{133}Ba that was added as a yield tracer just prior to microprecipitation. We were, however, able to recover 95% of 6 dpm ^{226}Ra added to 2- 5 L of NYC water samples measured by either LSA (without ^{133}Ba) or alpha spectrometry .